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Researchers simulate self-focusing beams in fiber

Improvements in the peak powers of rare-earth doped fiber lasers and amplifiers are now enabling these sources to replace conventional lasers in a variety of applications. Recent achievements of multi-megawatt pulses are approaching a fundamental limit imposed by self-focusing (SF) within the fiber core. Although fibers are generally fabricated from fused silica, which exhibits a relatively low increase in the refractive index (RI) with optical irradiance (power per unit area), the profile of the fundamental mode can induce a slight lens-like addition to the RI profile (see Figure 1). As the mode propagates, the lens causes the beam waist to contract, further increasing the irradiance. If the beam power exceeds a value defined as P_{crit}, catastrophic self-focusing will ultimately occur. At powers below P_{crit} the increased diffraction or spreading effect of the smaller beam waist overcomes the

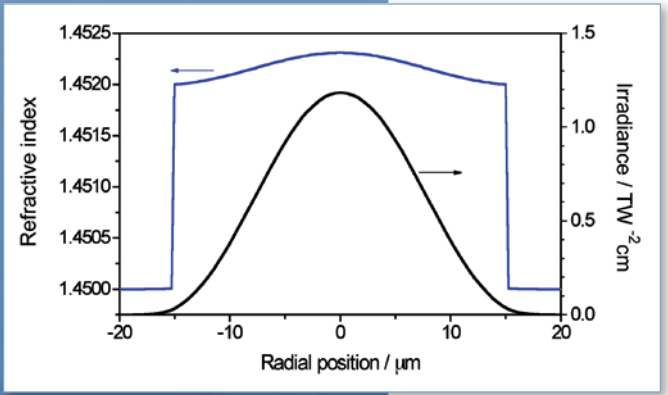

Figure 1. Refractive-index profile (RIP) and fundamental-mode irradiance profile calculated for a step-index large-mode-area (LMA) fiber at 3MW beam power. The bulge at the center of the RIP results from the nonlinear response of the refractive index to the very high irradiance (Kerr effect); it acts like a lens during propagation. (Continued on page 2)

Sandia National Laboratories

CRF scientists measure the extinction coefficient of soot

Measuring the amount of soot in flames and fires, calculating the amount of radiation emitted by hot soot, and predicting the shielding effects of smoke clouds are all fundamental to understanding the hazards associated with large fires. These measurements and calculations rely on accurate knowledge of the optical properties of soot and, in particular, its dimensionless extinction coefficient (K_e). Thanks to a team of Sandia scientists this important coefficient was recently quantified for soot extracted directly from the interior region of laminar diffusion flames, burning a range of fuels from methane to a JP-8 jet fuel surrogate. The measurements form part of the experimental database of sooty diffusion flames (see CRF News Sept/Oct 2005) and represent the first determination of "in-flame" soot K_e for laminar diffusion flames.

Figure 1. Laminar diffusion flames of ethylene (left), JP-8 surrogate (center) and methane (right) burning in air.

The research team for this work included Tim Williams and Chris Shaddix from the CRF and Jill Suo-Anttila and Kirk Jensen from Sandia's Fire Science and Technology department in Albuquerque, NM. Additional support was provided by Nancy Yang, who provided transmission electron micrograph (TEM) images of collected soot aggregates.

The researchers investigated three fuels burning in air: methane, ethylene, and a six-component JP-8 fuel surrogate developed at the University of Utah. The flames were supported on burners with coannular geometries. For the JP-8 surrogate flame, a capillary-driven ceramic vaporizer was used to flash-vaporize the fuel mixture at the burner base. Figure 1 demonstrates the different sooting propensities of the three fuels, with soot exiting the top of the JP-8 flame as smoke (despite the use of a significant amount of nitrogen added to the vaporized fuel to reduce the soot loading).

To measure K_e, researchers extracted soot through a small metal tube placed into the flame. After being diluted with nitrogen, the sample was

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Self-focusing beams (Continued from page 1)

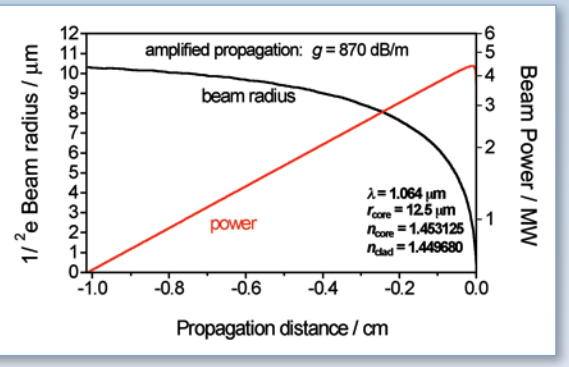


Figure 2. The fundamental mode of an LMA fiber amplifier was amplified from an initially low power source (500W; not shown) using a beam-propagation method (BPM) code. Beam radii and total power are shown as a function of distance from catastrophic self-focusing. The nonlinear coefficient was taken to be $n_2 = 2.7 \times 10\text{-}16$ cm^2 W^{-1} based on an average of published values.

self-focusing effect so that a zero beam waist is avoided, although oscillations in the waist may occur along the propagation direction. If not accounted for, such effects can strongly influence the output spectrum (due to enhanced nonlinear processes in the fiber), beam quality, and the damage threshold of the device.

It is clear that further power scaling will be significantly enhanced by accurate and reliable simulations of these effects, which will facilitate the design of high-power fiber amplifiers by reducing the experimental testing needed. While self-focusing is a well-understood phenomenon in bulk dielectrics, its effect on the propagation of guided modes in waveguides is subject to conflicting conclusions in the literature. For example, some authors of theoretical studies have claimed that a high-power mode propagating in a fiber will necessarily undergo large oscillations in waist size, which would substantially lower the threshold power for nonlinear processes and optical damage. Some have concluded that the SF limit is significantly different in a fiber than in a bulk dielectric identical to that of the fiber core, while others have predicted little difference. Since all previous calculations were performed for fibers without gain and fibers that were straight (bending or coiling significantly distorts the mode distributions), Sandia researchers in California and New Mexico, supported by the Fiber Laser Grand Challenge Laboratory Directed Research and Development (LDRD) set out to answer the following questions:

—How does self-focusing affect the propagation of beams undergoing amplification at powers approaching P_{crit}?
—How does coiling of the fiber affect propagation of amplified and unamplified beams at powers approaching P_{crit}?
—How does P_{crit} compare between straight and coiled step-index fibers and a similar bulk material?

Using a Sandia-developed beam propagation method (BPM) code, the researchers simulated amplification of the fundamental mode of a straight (uncoiled) step-index fiber, starting at a beam power of 500W where SF effects are negligible. The final cm of propagation before catastrophic self-focusing occurs is indicated by the plot of beam waist (radius at e^{-2} of peak power) and beam power, as shown in Figure 2. Note that the waist decreases smoothly as a result of SF and that no oscillations are observed. Figure 3 shows plots of the transverse profile of the amplified beam at various powers; the profile clearly narrows as a result of self-focusing as P approaches P_{crit}, previously found to be ≈ 4.3MW. We found that the transverse profile at any particular power would propagate nearly unchanged at that power in the absence of gain. In other words, the amplified profile represented a stable "mode" or solution which had adapted to SF. This remarkable result is illustrated by the solid blue curve in Figure 4 for the high-power solution at 3MW; only slight oscillations (believed to result from numerical approximations of the BPM) are observed. In comparison, the fundamental mode exhibits large oscillations when propagated at 3MW (solid red curve). As a further test of the uniqueness of the high-power solution, we used an eigenmode solver (a code that finds field distributions that propagate without changing shape) to find a solution using an iterative approach. The initially calculated fundamental eigenmode was used to compute nonlinear contributions to the waveguide at a power of 3MW. A new eigenmode was generated using this perturbed waveguide and the process was repeated until the mode profile converged. The resulting solution was then propagated at fixed power in the same manner as before, resulting in the beam-waist variation indicated by blue circles in Figure 4. A comparison of the profiles of the two solutions (not shown) showed no significant differences, demonstrating that a unique stable "mode" was obtained in both cases.

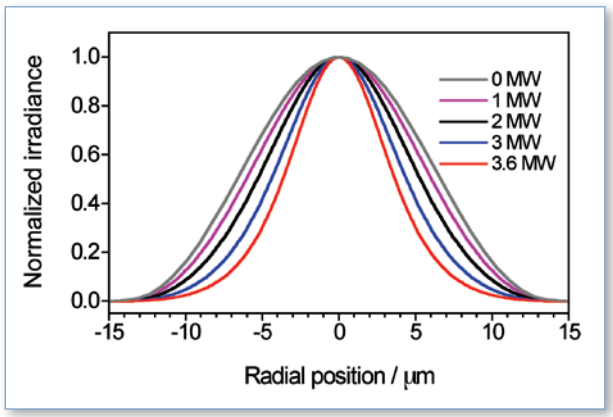


Figure 3. Comparison of transverse profiles of the amplified beam from Figure 2 at the indicated beam powers. The narrowing of the profile results from self-focusing, which becomes more significant with increasing power.

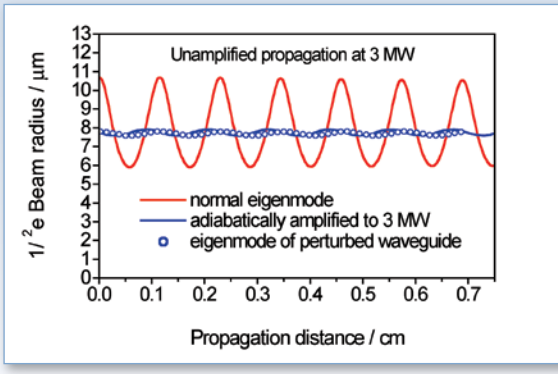


Figure 4. Comparison of unamplified propagation at 3MW of the following beams: the normal fundamental eigenmode of the waveguide (red curve), the eigenmode previously amplified to 3MW (blue curve; profile plotted in Figure 3), the eigenmode of the waveguide with nonlinear index contributions included, determined in an iterative calculation (blue circles).

A similar study performed with the fiber coiled to a bend radius of 0.84 cm resulted in the same conclusions, although the quantitative details were somewhat different.

In conclusion, the team found that at high powers, lower than P_{crit} (e.g., 3MW), non-oscillatory (stationary) solutions exist that propagate unchanged in step-index fibers. In a fiber amplifier seeded with the fundamental mode, the transverse spatial profile will evolve into that of the stationary solution as the beam is amplified, i.e., the beam will not undergo longitudinal oscillations.

These conclusions hold for coiled as well as straight fibers. For a given value of n_2, P_{crit} is nearly the same in the bulk material and in a step-index fiber, either straight or coiled.

Guanghua Wang, Mathew Dunn, Bassam Dally, Rob Barlow and Bob Harmon

Barlow lab hosts visitors

Along with Guanghua Wang and Bob Harmon, Rob Barlow is hosting two Australian visitors in the Turbulent Combustion Laboratory. Matthew Dunn, a Ph.D. student from the School of Mechanical, Mechatronic and Aeronautical Engineering at the University of Sydney, working under Professors Assaad Masri and Bob Bilger, brought a piloted premixed jet burner (PPJB) designed for the investigation of highly turbulent premixed methane/air flames. Bassam Dally, a Senior Lecturer at the School of Mechanical Engineering at the University of Adelaide, brought a different type of premixed burner. His project is concerned with the effects of turbulence intensity and reactant temperature on the preheat zone of flames with methane and ethylene mixtures. Dally has visited the CRF twice before, in 1995 and in 2001.

Valorani from Italy, Goussis from Greece at Najm lab

The laboratory of Habib Najm had two visitors for the month of October, Mauro Valorani and Dimitrios Goussis. They were involved in a multiyear collaboration with Najm to develop computational tools for analysis and reduction of chemical models of relevance in combustion. Dr. Valorani is a professor of aerospace propulsion at the Dipartmento di Meccanica e Aeronautica, Universita degli Studi di Roma "La Sapienza". Dr. Goussis will be taking up a new position as Associate Professor at the National Technical University of Athens.

CRF researchers recognized by SAE

John Dec and Dennis Siebers were recognized by the Society of Automotive Engineers (SAE) for organizing the 3rd SAE International Symposium on Homogeneous Charge Compression Ignition (HCCI) Engine Combustion and a laboratory tour of the CRF. The conference took place September 24 - 26, 2006, in San Ramon, CA. 136 people attended, including 43 international visitors from 10 countries.

Dec received a SAE Excellence in Oral Presentation Award for his presentation of a paper on thermal stratification in HCCI engines at the SAE International Congress.

Dec and co-inventor Carl-Magnus Sjöberg were also awarded a patent on Fuel Mixture Stratification as a Method for Improving Homogeneous Charge Compression Engine Operation on October 31, 2006.

Caroline Genzale works with engines group

Caroline Genzale, a Ph.D student in the Engine Research Center at the University of Wisconsin, is visiting the engines group for six months, working with Mark Musculus to perform engine experiments that will be used to validate computationally-derived combustion chamber designs for low temperature combustion.

Professor Andre Boehman, visits Advanced Fuels Lab

Professor André Boehman spent the summer conducting experiments with Sandians Chuck Mueller and Glen Martin using the optical engine in the CRF's Advanced Fuels Laboratory. Their work focused on elucidating the role of soot radiative heat transfer in the observed higher NO_x emissions when an engine is fueled with biodiesel, and on determining the extent to which the biodiesel NO_x increase is a function of the level of exhaust gas recirculation. Boehman is a professor of fuel science in the Department of Energy and Geo-Environmental Engineering at Pennsylvania State University.

Calculating rate coefficients from first principles

Reactions between two resonantly stabilized free radicals play an important role in the gas phase chemistry leading to the formation of polycyclic aromatic hydrocarbons and soot in flames burning aliphatic fuels. The theoretical prediction of rate coefficients and product distributions for such reactions is complicated by two factors. First, these reactions invariably take place over multiple, interconnected potential wells. Second, the initial complex-formation steps (and some dissociation steps) are "barrierless." Without a potential barrier, calculating these rate coefficients from first principles becomes a tricky problem.

Recently, Yuri Georgievski (Sandia), Stephen Klippenstein (now at Argonne National Laboratory), and Larry Harding (Argonne) developed quantitatively accurate methods for attacking this latter problem on the basis of variational transition state theory (TST). TST provides a powerful tool for calculating rate coefficients of chemical reactions. It assumes that there is a "bottleneck", called a transition state, in the configuration space between reactants and products through which the reactive trajectories pass only once. From a mathematical point of view, the transition state is simply a dividing surface that separates reactants from products and through which the probability flux is minimal. Usually, the transition state is situated at the top of the barrier separating reactants from products. For barrierless reactions, however, this kinetically important feature is absent and the transition state must be determined variationally. A typical "radius" of the optimal dividing surface for a barrierless combustion reaction may vary from 2-3 angstroms at 2000-2500K to 6-7 angstroms at room temperature and below.

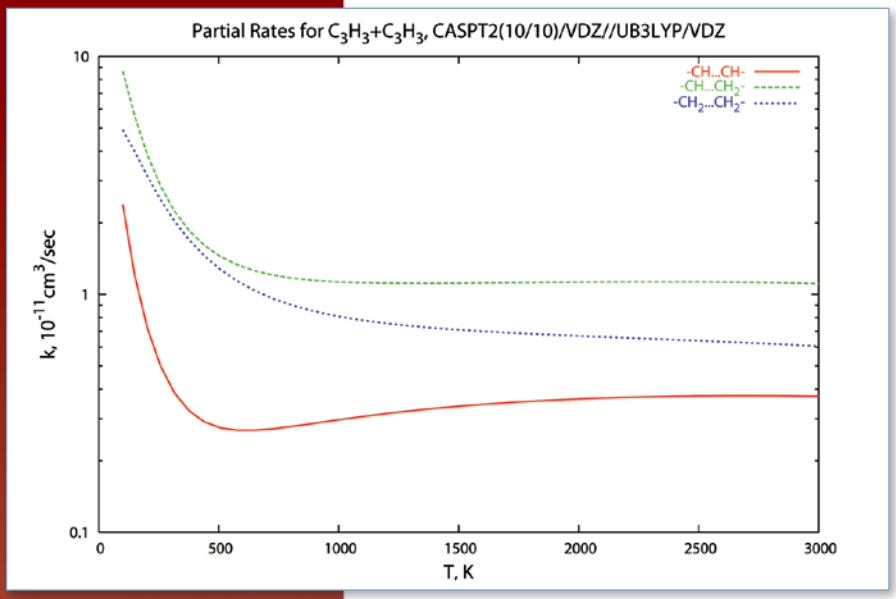


Figure 2. Rate coefficients for the head-tail (-CH...CH$_2$-), head-head (-CH...CH-), and tail-tail (-CH$_2$...CH$_2$-) channels of propargyl + propargyl reaction as functions of temperature.

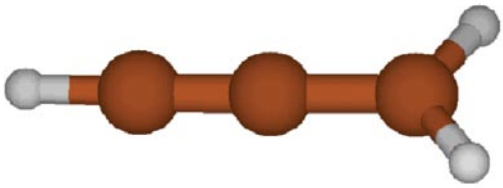


Figure 1. Propargyl radical

In the methodology developed by Georgievski and co-workers, the potential energy of interaction among the atoms is calculated "on the fly" with *ab initio* (first principle) quantum chemistry methods. This method enables calculations to be performed efficiently on a cluster of computers with multiple working nodes. For practical applications, it is important to be able to construct a small set of dividing surfaces that are sufficiently flexible to minimize the reactive flux over a broad range of temperatures. This problem is complicated by the fact that many important combustion reactions have multiple association channels that must be treated simultaneously. For example, the propargyl radical, (shown in Figure 1), which is a key species for soot formation, has two binding sites ("the head" and "the tail") associated with either of the terminal carbons. As a result, there are three distinct channels for the propargyl + propargyl association reaction: "head-head", "tail-tail", and "head-tail".

The method developed by Georgievski, *et al.* is based on the concept of a "multi-faceted dividing surface". The reactive flux through each facet represents the rate through a specific channel. Together, the facets form a closed dividing surface that satisfies the variational principle. The probability flux through this surface provides an upper boundary for the true rate coefficient. Thus, by minimizing the reactive flux through a set of dividing surfaces, one obtains the rate coefficient over a broad range of temperatures. Comparison with classical trajectory calculations for small systems shows that the accuracy of the method is in the range of 10%. The calculated association rate coefficients for the C_3H_3+C_3H_3 reaction for each channel are shown in Figure 2 as functions of temperature. This reaction leads ultimately to formation of the first aromatic ring, which is believed by some to be a rate-limiting step in soot formation. The calculated association rates compare favorably with experiment at low temperatures where experimental data are available. Using the so-obtained reactive fluxes in combination with those developed by Jim Miller (Sandia), master equation methodology will allow obtaining the rate coefficients for this important reaction *ab initio* in a broad range of temperatures and pressures and to permit using them for combustion modeling of soot formation.

Measuring soot

(Continued from page 1)

drawn into a 1m long tube where extinction measurements were made using laser beams and photodiode detectors. The soot volume fraction present in the tube was calculated by collecting the sampled soot on a filter and weighing it, and by combining this measurement with the extinction measurement, the dimensionless extinction coefficient could be determined.

At a wavelength of 635 nm the researchers found K_e for the ethylene and JP-8 flames to equal ~9 and ~10 respectively, which is consistent with K_e values of post-flame soot (smoke) reported in the literature.

However, K_e values of 9-10 are considerably higher than have traditionally been used by combustion researchers. K_e is the sum of the scattering and absorption coefficients ($K_e = K_s + K_a$) and it is often assumed that soot particles are small enough for the scattering component to be ignored, so that $K_e = K_a$. Based on this assumption, K_e can be calculated using Mie theory in the Rayleigh limit. Using this approach and commonly used refractive index values of soot at visible wavelengths, K_e values of 3-5 are calculated. When used to interpret soot volume fraction measurements, this traditional approach to estimating K_e produces apparent soot concentrations at a factor of two higher than the current K_e measurements imply.

Over the past decades it has become apparent that part of this discrepancy is due to the heavily aggregated nature of soot, which causes the scattering component of extinction to become significant. Figure 2 shows a Transmission Electron Microscope (TEM) image of a typical JP-8 soot aggregate with small, 40 nm diameter, soot primary particles joined together to make a much larger chain-like structure of several hundred nanometers in size. While individual soot primaries fall in the Rayleigh regime, the aggregate as a whole does not.

The importance of scattering to most K_e values is highlighted by considering the K_e result for the methane flame, which was measured by the research team to be ~7.0, significantly lower than was found for the other two flames. Inspection of the TEM images of methane soot aggregates showed much smaller aggregates containing only a few primary particles. Calculations suggest that scattering from these aggregates only amounts to between 1-3% of the total extinction. This lack of significant scattering conveniently accounts for the lower K_e value.

The research team used the results from the K_e measurements and the TEM images to speculate on a new value for the refractive index of soot. Assuming (1) the K_e value of methane soot is effectively a measure of the absorption coefficient such that $K_a = K_e$, and (2) the absorption coefficient of soot particles is relatively independent of fuel source (since all mature soot particles are primarily composed of carbon), it follows that $K_{a,\,\text{methane}} = K_{a,\,\text{ethylene}} = K_{a,\,\text{JP-8}} = 7.0$. Therefore, the percentage of soot scattering for soot from the ethylene and kerosene coannular flames is in the range of 20–30%. Using the Rayleigh-Debye-Gans (RDG-PFA) theory of scattering and absorption by polydisperse fractal aggregates to analyze the soot aggregates, the research team found solutions of the real and imaginary components of the refractive index of soot at 635 nm that are consistent with the current measurements. Figure 3 shows the results of such an analysis for the ethylene soot, with solutions formed at the intersection of ellipses for K_a and the scattering albedo, ω_a (= K_s/K_e). Several ellipses are plotted to mark the bounds of possible K_a and ω_a values. At $K_a = 7$ and $\omega_a = 0.25$, the complex refractive index of soot is $m = 1.98 - 1.66i$, which has an imaginary (absorption) component that is substantially larger than that of soot refractive index values quoted in the literature (graphite is the only carbon form with this sort of index). However, using the lower end of the scattering albedo implied from the K_e measurements, $\omega_a = 0.17$, yields a refractive index $m = 1.75 - 1.03i$ that is similar to a recently reported value for soot. These results are being reported in an upcoming edition of the International Journal of Heat and Mass Transfer.

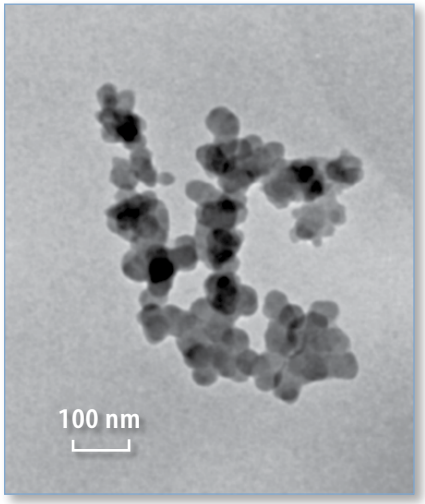


Figure 2. TEM image of a typical JP-8 soot aggregate.

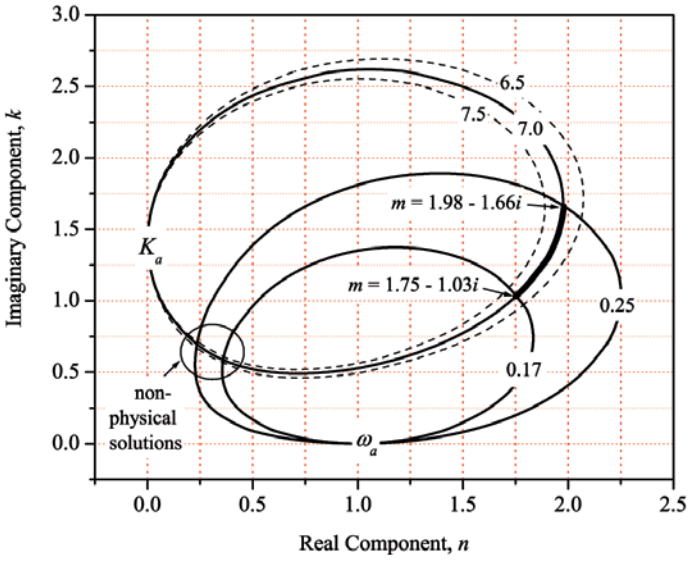


Figure 3. Solutions of the real and imaginary components of the refractive index of soot at 635 nm that are consistent with particular values of the nondimensional absorption coefficient, K_a, or the scattering albedo, ω_a. The intersections of the ellipses denote index values that match both the assumed K_a and ω_a values and are consistent with the current K_e measurements. Values of the real part of the refractive index less than one are considered nonphysical.

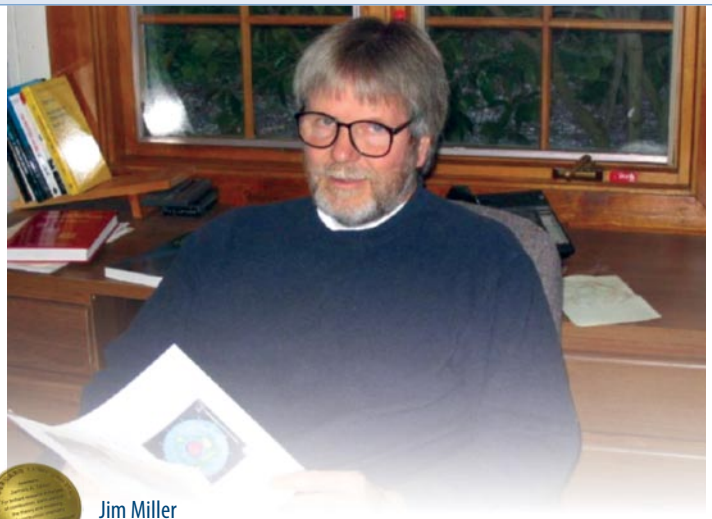
Jim Miller awarded gold medal

Crf scientist Jim Miller researches how pollutants are formed in combustion and devises chemical kinetic models to enable better emissions control. He has worked on the nitrogen chemistry of combustion (i.e. NO_x formation and control) and the gas-phase chemistry leading to soot formation. This August, at the Thirty-First International Combustion Symposium, held in Heidelberg, Germany, The Combustion Institute awarded Miller the Bernard Lewis Gold Medal "for brilliant research in the field of combustion, particularly on the theory and modeling of combustion chemistry".

Miller's paper "Mechanism and Modeling of Nitrogen Chemistry in Combustion" (J.A. Miller and C.T. Bowman, Prog. Energy Combust. Sci. 15, 287-338 (1989), is the single most cited paper ever to appear in any combustion journal. Miller's 1992 work, "Kinetic and Thermodynamic Issues in the Formation of Aromatic Compounds in Flames of Aliphatic Fuels" (J.A. Miller and C.F. Melius, Combust. Flame 91, 21-39 (1992) is the most cited paper to appear in the journal Combustion and Flame's 50-year history.

Together with Bob Kee, now at the Colorado School of Mines, Miller created and developed CHEMKIN, now the standard for chemical kinetic modeling in combustion. Along with Sandia collaborators, Miller and Kee developed mathematical and computational strategies employed in application codes such as PREMIX, PSR, SENKIN, OPPDIF, and CRESLAF. In 1990, Miller received the Silver Medal from The Combustion Institute for theoretical work on the structure and extinction of opposed-flow premixed flames (with Bob Kee, Graham Dixon-Lewis, and Greg Evans).

Miller is currently collaborating with Stephen Klippenstein, now at



Jim Miller

Argonne National Laboratory, on research involving the theoretical description, using master equations, of reactions that take place over multiple, interconnected potential wells and of the thermal dissociation of weakly bound free radicals. Such reactions dominate our understanding of all of combustion chemistry, as well as that of atmospheric chemistry and chemical vapor deposition.

Miller received his bachelor's degree from the University of Cincinnati and his Ph. D. from Cornell University in 1974. Part of the founding staff of the Combustion Research Facility in 1980, Miller has held the title "Distinguished Member of the Technical Staff" since 1989.

Miller is a Fellow of the American Physical Society, a Fellow of the the American Association for the Advancement of Science, a member of the American Chemical Society, and The Combustion Institute. A special issue of The Journal of Physical Chemistry A, to appear in early 2007, will be devoted to a James A. Miller Festschrift in celebration of his 60th birthday.

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